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Oscillatory behavior of the magnetic moments of gold-covered iron surfaces

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Local magnetic moments at iron (001) surfaces have been studied by electronic structure calculations employing the slab geometry up to 17 layers. Whereas the clean Fe(001) surface shows oscillations in the local magnetic moments, no oscillations were observed in the case of coverage by a monolayer of gold, in sharp contrast with the gold/iron multilayer system, where they reappear. The screening effect of the oscillatory behavior on Fe(001) surfaces is very local: Fe(001) surface covered with a submonolayer of gold shows oscillations for the noncovered parts of the surfaces. [S0163-1829(98)02136-5]

INTRODUCTION

The electronic structure of the iron (001) surface has been studied quite extensively, both experimentally and theoretically.¹⁻⁴ Of special interest are the deviations of the total magnetic moments at the surfaces as compared with the bulk. Early experiments showed a magnetic “dead” surface,^{5,6} in contradiction with band structure calculations, which showed an enhancement of the magnetic moment at the surface.^{7,8} More recently, photoelectron spectra⁹ and Mössbauer experiments¹⁰ confirmed the enhancement of the moments at the surface.

One of the noble metals that matches very well on the Fe(001) is gold. Since it forms no solid solutions with iron the Au-Fe system has been studied extensively.^{11,12} Also, gold layers are being used to protect iron from oxidation. Application of iron in a (spin-resolved) scanning tunnel microscope is actively being investigated.^{3,4,13} In view of this, the magnetic properties of the Fe(001) clean surface and gold covered deserve attention.

Another important question is how the surface properties evolve into the bulk behavior as a function of the distance from the surface. Wang *et al.* found strong oscillatory behaviors for both charge and magnetic moments for the clean Fe(001) surface.^{1,7} Subsequently, Ohnishi *et al.* found no charge oscillations and reduced magnetic moment oscillations, which was interpreted to arise from the slab approximation.^{8,14}

DETAILS OF THE CALCULATIONS

Ab initio band structure calculations were performed in the slab supercell approach with the localized spherical wave method.¹⁵ Since the magnetic properties are sensitive to the Brillouin zone sampling, a dense mesh of 1 *k*-point per 10^{-6} Å³ was employed. In order to allow comparison of charge and moments, Wigner-Seitz radii that deviate negligibly from the bulk values were employed. A series of calculations were performed on the clean iron (001) surface in order to investigate the influence of the slab approximation on the magnetic properties. These comprised of 11 mono-

layer (ML) of iron with 9 ML of vacuum (empty spheres) (11+9) as well as (7+5) and (5+7) systems. The influence of the vacuum layer was studied on the system of 11 ML of iron with interslab distances from 2.2–14.8 Å. The gold covered Fe(001) was studied on a system with 17 ML iron, 2 ML gold, and 9 ML vacuum, with gold in the (110) orientation. The partial gold coverage was simulated by doubling the cell in one or two directions perpendicular to the surface direction with only one of the gold position occupied. A gold/iron multilayer was calculated as reference consisting of 11 ML Au and 9 ML Fe.

CLEAN IRON (001) SURFACE

First the convergence as a function of the thickness of the iron slab is considered. In Fig. 1 the magnetic moments and the charge for majority and minority Fe 3*d* states are given as a function of depth from the surface for the (11+9) and the moments for the (7+5) system. Although the outermost iron layers are remarkably identical the innermost Fe layers in the case of the (7+5) system deviate clearly from the bulk iron. The central layers in the case of the (11+9) calculations

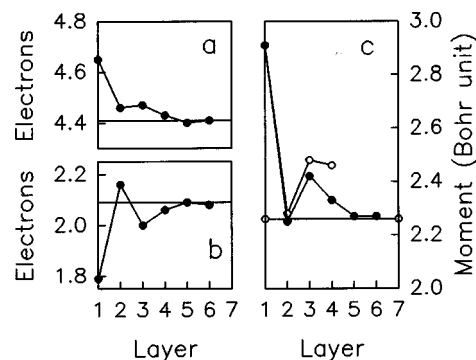


FIG. 1. Number of Fe 3*d* electrons for the majority-spin direction (a) and minority-spin direction (b). (c) Magnetic moments from the surface to the center for the (11+9) (filled circles) and (7+5) system (open circles).

approximate bulk iron much better. This leads to the conclusion that the surface electronic structure is less sensitive to the slab approximation than the bulk properties. The magnetic moments converge slower to the bulk value than the charge: whereas the charge is reasonably converged to the bulk value from the third layer, the magnetic moment reaches the bulk value from the fifth layer. In order to test the influence of the thickness of the vacuum slab in between the iron slabs a series of calculations were performed with the interslab distances between 2.2–14.5 Å. With increasing interslab distances the magnetic moment of the surface iron layer increases until a maximum is reached of $2.95 \mu_B$ at 2.87 Å. Beyond this distance the moment drops to a value of $2.91 \mu_B$ at 6 Å and remains constant beyond. Unless stated differently, the remainder of this report is based on the results for the (11+9) system.

The outermost iron layer loses about 0.50 electrons from mainly the delocalized $4s$ and $4p$ states: 0.32 electrons are transferred to the vacuum as compared with the bulk and 0.17 electrons are donated to the subsurface layer, which is negatively charged. A strong oscillatory behavior is found for the magnetic moments, originating from an additional charge transfer from the minority $3d$ to the majority $3d$ states. The combined result is that the major contribution to the oscillatory behavior of the magnetic moments is from the minority electrons. The oscillatory behavior for the (11+9) and the (7+5) systems is identical for the two outermost layers but deviate quantitatively for the more inner layers.

The surface iron layer shows a reduced width of the bands with a somewhat increased exchanged splitting. Consequently, the majority d band is almost filled and the system approaches the polarization situation of a strong magnet like Co at the surface. Accordingly, the polarization of the conduction electrons at the surface is reversed with respect to the bulk.

Fe(001) WITH A MONOLAYER COVERAGE OF GOLD

The gold monolayer covered Fe(001) showed a slower convergence towards bulk properties as compared with the uncovered system. For this reason the thickness of the slab was increased to 19 ML. A significant difference is found in the outermost iron layers, which are practically neutral (7.98 and 8.06 electrons for the surface and subsurface layer, respectively), while the covering gold atom loses 0.30 electrons towards the vacuum. This directly influence the behavior of the magnetic moments as a function of depth (Fig. 2). A rather smooth decrease of the magnetic moments is found moving from the surface towards the bulk. The net magnetic moment of the gold is small: $0.07 \mu_B$. These data were obtained with the experimentally determined Fe-Au distance at 2.68 Å,¹⁶ and proved to be quite sensitive to the Fe-Au distance: a calculation with 2.87 Å (about the Au-Au distance) showed a reduction of the magnetic moment on Au to $-0.0005 \mu_B$, an increase in the magnetic moment of the outermost iron layer to $2.65 \mu_B$, and the reappearance of a (weak) oscillatory behavior in the magnetic moments as a function of depth.

The density of states at the Fermi level for the surface Fe atoms differ much from the bulk-like Fe atoms, as in the case of the clean Fe(001) surface. The partial Fe $3d$ density of

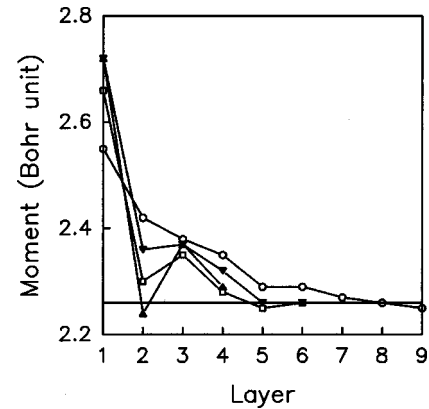


FIG. 2. Layer-resolved magnetic moments of iron from the surface to the center for (1) Fe (001) surface covered by a monolayer Au (open circles), (2) Fe (001) surface covered by half a monolayer Au (triangles down: under gold covered part, and triangles up: uncovered part), and (3) Fe-Au multilayer system (squares).

states at the Fermi level increases steadily from surface to inner layer for the spin-up electrons, while it decreases rapidly from the surface to inner layers for the spin-down electrons, as shown from Fig. 3.

Fe(001) SURFACE WITH SUBMONOLAYER Au COVERAGE

The doubling of the unit cell in one direction perpendicular to the surface with only one gold position occupied leads to a structure with one type of iron at the outermost layer, two types of iron (one directly under the Au atom, the other under a vacuum position) at the subsurface, one type of iron at the next layer, etc. The behavior of the magnetic moments as a function of depth is shown in Fig. 2. The outermost iron atom shows a moment of $2.72 \mu_B$, in between the clean Fe(001) ($2.91 \mu_B$) and the monolayer Au covered Fe(001) ($2.55 \mu_B$). The behavior of the Fe atoms in the next layer is very different: the iron under the gold atom shows a moderate reduction in the magnetic moment, whereas the iron atom under the unoccupied part of the surface shows a strong reduction in the moment, resulting in a value even smaller than the bulk value. The iron in the next layer is enhanced again ($2.35 \mu_B$). Thus, the suppression of the oscillation of the magnetic moments by the gold at the Fe(001) surface is a

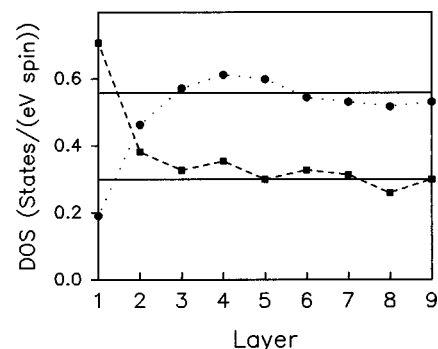


FIG. 3. Layer-resolved density of states at Fermi energy for spin-up (circles) and spin-down (squares) electrons for the Au monolayer covered Fe (001).

very local phenomenon: the uncovered part of the irons still shows oscillations as a function of the distance from the surface, while for the covered part the oscillations have practically disappeared. A similar phenomenon is found in the Fe(001) surface covered by a quarter monolayer of gold. The top iron layer shows a moment of $2.84 \mu_B$. The magnetic moments for the next-to-top layer iron atoms are $2.39 \mu_B$ (iron atom under gold), $2.44 \mu_B$ [iron in the (001) direction] and $2.30 \mu_B$ [iron in the (110) direction].

GOLD-IRON MULTILAYER SYSTEM

An interesting question arises: how do the magnetic moments behave in a gold-iron multilayer system, where the gold layer in contact with the iron will not lose charge to the vacuum? This question was examined in a calculation on a system with 11 ML Fe, 9 ML Au. Results are displayed in Fig. 2. There is a charge transfer of 0.15 electrons from the interface Fe layer to the interface gold layer, in contrast with the monolayer gold on the Fe(001) surface case. Both charge and moments show Friedel-like oscillations, in contrast with the Au monolayer covered Fe(001) surface case.

CONCLUSIONS

In case of the clean iron (001) surface, the outermost iron layer loses about 0.50 electrons because of the tails of the delocalized *s* and *p* functions extending into the vacuum. As a function of depth an oscillatory behavior of both spin and charge results. By analogy, in the case of the monolayer coverage by gold one expects the outermost iron layer (being the second outermost layer from the surface) to adopt a negative charge, but this effect is compensated by an opposite charge transfer to the much more electronegative gold. Hence, no oscillatory behavior is found. In the case of a gold-iron multilayer a charge transfer to the gold occurs and, hence, the oscillatory behavior of the magnetic moments reappears. The suppression of the oscillatory behavior by gold coverage is very local: in the case of a partially covered Fe(001) surface, the oscillations are suppressed under the covered part of the surface only. For all the cases a reversal of the polarization of the density of states at the Fermi energy occurs.

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